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The Beta-Ray Spectra of Cu^{64} at Low Energies

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An electrostatic beta-ray spectrometer has been constructed utilizing the focusing action of the electric field between concentric cylindrical conductors. A Geiger tube counter using an argon-alcohol mixture was used; a technique for making windows for the counter was developed whereby electrons of energies as low as 5 kev could be counted. A source of radioactive Cu^{64} was mounted on a very thin collodion film in order to eliminate the effects of back scattering. The positron and electron spectrum of Cu^{64} was plotted with the apparatus in the region below 50 kev. It was found that the ratio of the number of positrons to the number of electrons in this region did not agree with the number predicted by the Fermi theory of beta-disintegration.

INTRODUCTION

THE continuous beta-ray spectrum has been the object of much investigation since its discovery. Many of the results have been discordant and are only lately beginning to show some agreement. Practically all the research has been confined to the upper region of the spectrum, the region below about 50 kev remaining completely unknown until quite recently. The object of the present research was the investigation of the electron and positron spectrum of Cu^{64} in the region below 50 kev.

Flammersfeld,[1] using RaE in a magnetic spectrometer, obtained the first reliable data in the low energy region. By comparing the curves obtained with sources mounted on supports of various materials and thicknesses, he was able to determine at what point the spectrum became distorted by the low energy electrons scattered back from the support. By using as a support a 0.1μ lacquer film, he obtained the spectrum down to 25 kev free from back-scattering. His work showed that in order to obtain the true spectrum at low energies, it is absolutely essential to use a very thin source and support. In fact, the work of Tyler[2] on the spectrum of Cu^{64} showed that the influence of back scattering was appreciable even at energies in the neighborhood of 200 kev; spectra obtained with thick sources showed a greater preponderance of electrons in the region below 400 kev than spectra obtained with thinner sources. Tyler did not attempt to measure the spectrum below about 60 kev.

For the present work it was decided to measure the two spectra of Cu^{64} in the extreme low energy region below 50 kev. Radioactive Cu^{64} was first investigated by Van Voorhis,[3] using a cloud chamber. The half-life is 12.8 hr.; it decays either by positron emission to Ni or by electron

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[1] A. Flammersfeld, Zeits. f. Physik 112, 727 (1939).

[2] A. W. Tyler, Phys. Rev. 56, 125 (1939).

[3] S. N. Van Voorhis, Phys. Rev. 50, 895 (1936).

emission to Zn. The ratio of the number of electrons to the number of positrons is about 1.6:1. The upper limits given by Tyler[2] for the electrons and positrons are respectively 0.578 $\pm$0.003 Mev and 0.659$\pm$0.003 Mev.

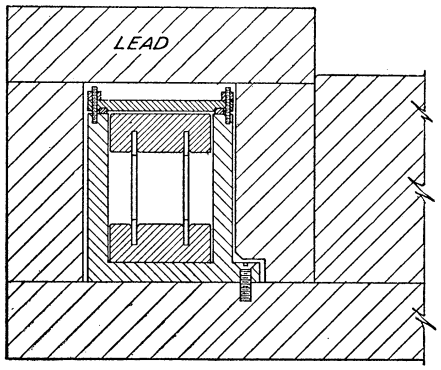

FIG. 1. Vertical section through spectrometer.

Since Cu^{64} emits both particles in nearly equal numbers, it is possible to plot both spectra under nearly identical conditions, using the same apparatus and the same source. From the formula given by Fermi[4] in his theory of beta-disintegration, the theoretical distributions and their ratio can be plotted and compared with the experimental curves.

APPARATUS

The apparatus used was an electric spectrometer originally constructed for work with recoil ions; it utilizes the focusing action of a cylindrical electric field, as discussed by Hughes and Rojansky.[5] Suppose two coaxial cylindrical electrodes of radii $\rho-\frac{1}{2}\Delta\rho$, $\rho+\frac{1}{2}\Delta\rho$ are maintained at a difference of potential ΔV. A charged particle of energy E will then travel along the median circle of radius ρ provided

$$\Delta V/E=2\log[(\rho+\tfrac{1}{2}\Delta\rho)(\rho-\tfrac{1}{2}\Delta\rho)]. \quad (1)$$

For any practical case

$$\Delta V/E=2\Delta\rho/\rho, \quad (2)$$

this expression being good to within 1 percent for $\Delta\rho<\frac{1}{3}\rho$. Suppose an entrance slit were located at the radius ρ; if now two particles enter the field through this slit with energy E and with their directions of motion making angles $+\alpha$, $-\alpha$ with the median circle, their paths will cross again at an angle

$$\theta=\pi/\sqrt{2}=127^\circ\ 17' \quad (3)$$

measured from the entrance slit. The point of crossing will be displaced toward the center by an amount $\rho\alpha^2/3$. If an exit slit of width ds is placed at the focus, the spread in energy of the particles passing through it is given by

$$dE/E=ds/\rho. \quad (4)$$

If $\phi(E)$ is the number of particles passing through the slit obtained as a function of energy E with the spectrometer, the true energy distribution $f(E)$ will be given by

$$f(E)=\phi(E)/E. \quad (5)$$

The vacuum chamber for the spectrometer was a portion of a ring-shaped trough of brass subtending an angle of about 120 degrees. It was turned from a brass casting, and it was found necessary to coat the outside completely with solder to make it vacuum tight. The inside measured $1\frac{5}{16}''$ wide by 2″ deep. The deflecting grids were spaced 0.62″ apart by means of slotted hard rubber blocks which also served to fix the grids in the center of the trough. The radius to the midpoint between the grids was 6.0″. Plates at the two ends of the trough carried adjustable rectangular openings to serve as entrance and exit slits.

The radioactive source was mounted on a rod which entered the apparatus through a sylphon bellows so that the position of the source could be adjusted. It was placed $\frac{1}{2}''$ in front of the entrance slit which was about $\frac{5}{32}''$ wide. A small gate operated from the outside through a Wilson type vacuum seal allowed the source to be closed off from the system so that background readings could be taken.

A small box was soldered on the other end of the trough outside the exit slit; this box carried the thin-window counter. To count electrons, the counter was mounted on the end of the box directly opposite the exit slit. The apparatus could also be arranged to count positive ions. To do this, the counter was mounted on the side of the box, and an insulated metal plate was

[4] E. Fermi, Zeits. f. Physik **88**, 161 (1934).
[5] A. L. Hughes and V. Rojansky, Phys. Rev. **34**, 284 (1929).

placed in the box in line with the exit slit and facing the counter window. When this plate is given a potential of -5000 volts, positive ions coming through the slit are accelerated toward the plate, where they strike it and liberate secondary electrons; these electrons are accelerated away from the plate, and since they leave perpendicular to it, then enter the counter through the thin window.

A vertical section through the apparatus is shown in Fig. 1, a plan view in Fig. 2. The apparatus is surrounded with lead as shown in order to shield the counter from gamma-rays from the source. It is evacuated by means of a two-stage oil diffusion pump backed by a Cenco Hyvac mechanical pump.

The apparatus was first tested with lithium ions obtained from a filament coated with Li_2CO_3. The deflecting plates first used were solid copper plates spaced 1″ apart; with these plates, the curve of counts *vs.* voltage on deflecting plates was very broad. By using a very narrow entrance slit and mounting the Li ion source about three inches away so as to get a narrow beam, double peaks were obtained, indicating that the ions were being reflected from the plates so as to pass through the exit slit at deflecting voltages other than the proper value. The solid plates were therefore removed and replaced by grids made of 7-mil wire wound on metal frames with about 0.05″ spacing between turns. With these grids a sharp peak was obtained at the proper voltage, but in some cases as many as four additional peaks could be counted, all at voltages across the grids higher than the proper value. These peaks were due to ions passing through the inside grid into the region between the grid and the chamber where the field was in the direction to bend them back out. It was found that by placing defining mica slits between the grids and mica baffles between the walls of the chamber and the grids, the additional peaks were removed and the apparatus then worked as expected.

THE THIN WINDOW COUNTER

The counter used was made by drilling a $\frac{5}{8}$″ hole through a 1″ square copper block $2\frac{1}{4}$″ long. Glass plugs waxed into the hole at either end carried a 0.010″ tungsten wire. Brass angles hard soldered to the sides of the block allowed it to be fastened to the spectrometer. A hole in the side adjacent to the spectrometer carried the grid which supported the collodion windows; this grid contained a hexagonal array of 397 holes, 0.013″ diameter, spaced 0.020″ apart. Rubber gaskets between the grid and counter and between the grid and spectrometer formed the vacuum seals. Figure 2 shows the counter as mounted for counting electrons.

The collodion windows are prepared as follows: Flexible collodion (e.g., Merck) is diluted with an equal volume of amyl acetate. Two drops only of this solution are dropped in rapid succession from an ordinary medicine dropper into a pan of distilled water. The pan should be at least ten inches in diameter. The solution spreads out over a circle some eight inches in diameter and dries in a few seconds; it can then be cut into quarters by sawing a wire through it with a rapid up-and-down motion, starting at the center. The quarters are lifted out by means of a wire frame shaped like Fig. 3. The frame is

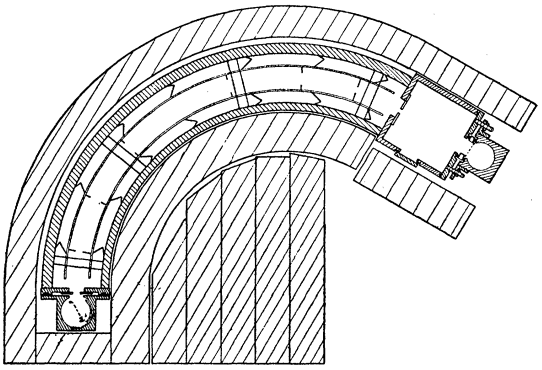

FIG. 2. Horizontal section through spectrometer.

placed under the center of the portion of film and lifted straight up so that the film drapes over the sides, thus forming a double layer. By this method breakage due to the surface tension of the water is minimized, as is also the occurrence of small pinholes. Interferometer measurements show that films formed in this way are about 0.08μ thick. Two such films placed in contact and observed in white light show a brilliant golden color.

The films when dry are given a very thin coat of silver by evaporation. A layer just visible as a

slight darkening is sufficient; such a layer is about 40 atoms thick. This silvering process is necessary to make the counter work properly. Counters used with unsilvered windows behaved in a very erratic manner. The grid upon which the film is mounted is dipped into acetone containing a few drops of collodion and dried. The film is then lowered on to it, silvered side up, and the wire frame cut away by means of a wire dipped in amyl acetate. The pressure of the gas in the counter is sufficient to seal the film vacuum-tight to the grid. No increase in pressure in the spectrometer could be noticed when the counter was filled. Figure 4 shows a curve of the transmission of the window for electrons plotted against the voltage of the electrons. It is seen that 4000 volts is sufficient for practically 100 percent penetration.

The counter is filled with a mixture of one volume ethyl alcohol vapor to about six volumes argon, the pressure used being 5 cm. The windows described above will stand about twice this pressure and will stand repeated fillings. The mixture should stand long enough for the two gases to mix thoroughly before the counter is used; filling the counter from a flask containing the mixture already prepared is the best way to accomplish this. The mixture will slowly diffuse through the counter-window, so a ballast flask is necessary to keep the pressure from falling too rapidly. The counter described was used with a ballast flask of 500 cc capacity, and one filling would last about eight hours, the counter voltage dropping about 200 volts during this time.

The counter was used with a high voltage supply of the type described by Gingrich[6] and the quenching circuit of Neher and Harper[7] feeding into a scale of 16 counting circuit. The counter used had a threshold of 800 volts when freshly filled and a plateau of 100 volts.

FIG. 3. Wire frame for lifting thin collodion films from the water surface.

[6] N. S. Gingrich, Rev. Sci. Inst. **7**, 207 (1936).
[7] H. V. Neher and W. W. Harper, Phys. Rev. **49**, 940 (1936).

PREPARATION OF THE SOURCE

The radioactive Cu^{64} for the source was obtained by bombarding zinc with deuterons from the 60″ cyclotron in the Crocker Radiation Laboratory, the reaction being $Zn^{66}(d, \alpha)Cu^{64}$. The copper was extracted by the electrolytic method described by Steigman.[8] The zinc is dissolved in HCl; 40 cc H_2SO_4 are added and the mixture boiled until SO_3 fumes are given off. This process is necessary to remove HCl from the solution, as copper does not plate well out of solutions containing it. The mixture is then diluted to about 300 cc and electrolyzed by use of a rotating cathode. The cathode used was a tungsten disk 2 cm in diameter, rotating 6000 r.p.m. Tests with solutions containing a known amount of copper showed that at a current of 10 ma, 2.9 v, two-thirds of the copper plated out in about three hours running. A drop of dilute nitric acid placed on the disk dissolved off the copper coating; the drop of solution was then picked up in a capillary tube and transferred to the collodion film support. This support was a 0.03μ film prepared as described above, using one drop of a more dilute collodion solution. A rectangular wire frame 1×3 cm carried the film. The drop of solution on its support was dried in a small vacuum chamber built for the purpose, then placed in the spectrometer. The effective width of the source was made small by tilting the film as shown in Fig. 2.

A voltage doubler circuit capable of giving 5000 volts above and below ground was used as the source of deflecting potential. From Eq. (2) above, $\Delta\rho/\rho=\frac{1}{10}$, $\Delta V=10{,}000$, so the highest energy beta-rays usable are 50 kev.

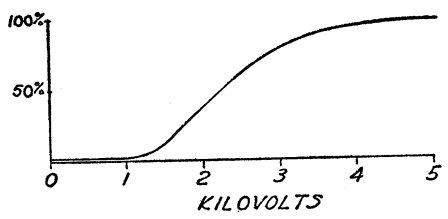

FIG. 4. Transmission of 0.1μ collodion counter window for electrons.

[8] J. Steigman, Phys. Rev. **53**, 771 (1938).

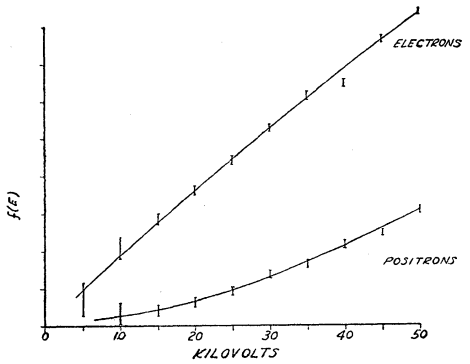

FIG. 5. Energy distribution of positrons and electrons from Cu^{64} below 50 kev.

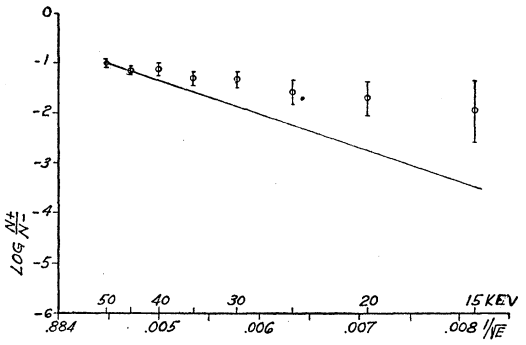

FIG. 6. Comparison of the theoretical and experimental ratio N_+/N_-.

RESULTS

Measurements were made on a source of about 15 mc. Background readings were taken periodically by closing the gate at the entrance slit. It was found that the background was independent of the voltage on the deflecting grids, being about 30 counts per minute. The highest counting rate measured was 1300 counts per minute, well within the capacity of the counter set. Counts were taken for two-minute periods and corrected for the decay of the source; the curves obtained plotted as a function of E were divided at each point by E to get the true distribution in energy of the positrons and electrons. The final curves obtained are shown in Fig. 5.

In 1934 Fermi[4] advanced a theory of beta-decay and gave a formula to represent the distribution of beta-rays in the continuous spectrum. From this formula the ratio of positrons to electrons in the Cu^{64} spectrum can be expressed as

$$N_+/N_-=A \exp\left[-2\pi\alpha Z((1+\eta^2)/\eta)^{\frac{1}{2}}\right], \quad (6)$$

where $\alpha=1/137$, Z the atomic number of the nucleus, and η the momentum of the particle in units of mc. A modification of the Fermi theory proposed by Konopinski and Uhlenbeck[9] gives the same expression for the ratio N_+/N_-. Using $Z=29$ and expressing η in terms of the energy E of the emitted particle in volts, one may for the region below 50 kev write Eq. (6) as follows:

$$\log N_+/N_-=k-670/(E)^{\frac{1}{2}}. \quad (7)$$

In Fig. 6 the straight line is the plot of Eq. (7), that is $\log N_+/N_-$ plotted against $1/(E)^{\frac{1}{2}}$. The circles represent the experimental values of $\log N_+/N_-$ taken from the original data. The constant in (7) has been determined so as to make the two curves coincide at 50 kev. The agreement is not good; the experiment shows a greater ratio of positrons to electrons at the low energy end of the spectrum than the theory predicts. This discrepancy cannot be ascribed to scattering due to the finite thickness of the source. This would change the form of the two curves of Fig. 5 but not their ratio, the positrons and electrons of a given energy being scattered equally.

In conclusion, I wish to thank Professor L. Alvarez for his interest and suggestions during the course of this work; Professor E. O. Lawrence for the use of the 60″ cyclotron in obtaining the radioactive copper used; Professor R. Oppenheimer and Dr. Philip Morrison for their theoretical contributions; and the entire staff of the Radiation Laboratory for their cooperation.

[9] E. J. Konopinski and G. E. Uhlenbeck, Phys. Rev. **48**, 7 (1935).